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# Release of radionuclides from waste rock and tailings at a former pilot uranium mine in eastern Finland

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The Paukkajanvaara mine in Eno, eastern Finland, started to operate in 1959. The mine was a test site for assessing the feasibility of larger-scale uranium extraction. Pilot-scale mining and milling were carried out between 1958 and 1961, and the mine site was rehabilitated in the early 1990s. The aim of this study was to examine the potential for further mobilization of radionuclides after remediation. There are two primary sources of contamination at the site, the waste-rock pile and the tailings. Our study indicates that $^{226}$Ra leached from the waste-rock pile and accumulated in surrounding soil. In run-off sediment samples collected from a dry stream bed near the waste-rock pile, the activity concentrations of $^{226}$Ra and $^{238}$U were higher than in soil samples. From the tailings, radionuclides can leach directly to the lake and to another small stream, which flows to the east of the waste-rock pile. In the water samples collected from this stream, uranium concentrations increased at the outlet to the lake. The results from the soil samples collected between the tailings area and the stream indicate leaching of $^{238}$U and $^{226}$Ra with the surface flow. Sediment samples collected from the bottom of the lake displayed pronounced uranium series disequilibrium with fractionations of $^{210}$Pb and $^{226}$Ra relative to the parent $^{238}$U. The results therefore indicate that leaching and accumulation of at least $^{226}$Ra from the waste-rock pile and possibly tailings is still ongoing.

## Introduction

Uranium mining and ore processing are known to be harmful environment and human health if the waste generated is not managed properly. Tailings contain trace elements including radionuclides that affect the metabolism of living organisms and can be a threat to human health (Jenne 1998, Sharma and Agrawal 2005). The main factors affecting the release of contaminants are the composition, volume and flow rate of the leaching solution (Benes *et al*. 1984). Owing to mobilization mechanisms, such as mineral dissolution, desorption and dispersion

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**Fig. 1**. Location of the former pilot-scale uranium mine at Paukkajanvaara in eastern Finland.

processes, radionuclides may migrate from tailings and contaminate nearby watercourses (Davé *et al*. 1982). Depending on their bioavailability, radionuclides may accumulate and become further concentrated through the food chain (Darko *et al*. 2009). A critical evaluation of mine remediation techniques is essential for determining their effectiveness and the long-term impact on the surrounding environment. This requires post-implementation data from samples taken well after the mine closure. (Lottermoser and Ashley 2005).

The Paukkajanvaara uranium deposit is located approximately 40 km north east of the town of Joensuu (Fig. 1). Pilot-scale mining and milling were carried out at Paukkajanvaara from 1958 to 1961, with 40 000 tonnes of ore at an average grade of 0.14% U treated in 1960–1961 (Äikäs 2012). Mining comprised both open-pit and underground operations, with mineral processing carried out on site. The ore was processed by crushing, grinding and leaching with sulphuric acid, followed by precipitation of uranium using sodium hydroxide or lime.

Ore processing generated tailings in the form of slurry waste, which was placed in a small pond adjacent to the mine without engineered barriers (Räisänen 1961). The pond was eventually filled with waste. Approximately 12 000 $m^3$ of fine-grained tailings and 7300 $m^3$ of waste rock were left on site (Mustonen *et al*. 1989). The area remained untouched for nearly 30 years until in the early 1990s, the site was remediated by covering the waste-rock pile and tailings first with 30 cm of clay and then 1.2 m of till. The Finnish Radiation and Nuclear Safety Authority (STUK) performed a radiological survey of the site both prior to and after rehabilitation. Following a final inspection in 1994, the area was deemed suitable outdoor use without restrictions (Colpaert 2006).

The aim of this study was to examine the potential for further mobilization of radionuclides from the tailings and waste-rock pile at Paukkajanvaara some fifteen years after remedial action was completed. Samples taken from the mining site included the following: (1) soil close to the waste-rock pile and tailings; (2) water samples from streams crossing the site and from the adjacent lake; (3) run-off sediments along a transect from the waste-rock pile to the lake; and (4) depth-profiled bottom sediment from the lake and samples of the tailings themselves. The results may help to estimate radiation doses to the local population as well as to improve disposal practices for uranium mill tailings.

## Sampling and analytical methods

### Site description

Paukkajanvaara is located in eastern Finland, approximately 40 km NE of Joensuu (Fig. 1, N 6981372, E 653304, ETRS-TM35FIN). The region belongs to the southern-boreal vegetation zone. The climate is temperate: in Joensuu, July and February mean temperatures are 16 °C and –9 °C, respectively. The mean annual precipitation is ca. 600 mm.

The Paukkajanvaara uranium deposit is located at an unconformity between Archean

basement rocks and Paleoproterozoic metasediments, which are intruded by mafic dikes (Piirainen 1968). The uranium ore bodies are located at the contact between a mafic dike and its country rocks; the latter are composed of quartzite, metaconglomerate, sericite-quartz schist, and Archean basement gneisses. The deposit consists of two main ore bodies, Mårtensson and Kunnansuo, and the main uranium ore minerals are uraninite and secondary uranophane (Makkonen *et al*. 1988). In addition, brannerite and coffinite are present as accessory uranium minerals (Makkonen *et al*. 1988). Uranium minerals occur mainly as fracture fillings, and uranium deposition post-dates the emplacement of the mafic dikes. The ore also contains magnetite, hematite, and galena with minor iron sulfides and chalcopyrite (Piirainen 1968).

## Sampling and sample preparation

The primary aim was to study the potential release of radionuclides from two sources at the site, the waste-rock pile (location 1 in Fig. 2) and the mill tailings (location 2). To study releases from the waste-rock pile, soil samples (location 3) and run-off sediment samples (location 4) were taken from its southeastern edge, where a small stream begins (location 5), being the discharge pathway for aqueous releases from the pile. Water samples were collected from this stream, which combines with another stream (location 6) not affected by the waste-rock pile and then flows into Iso Hiislampi (location 7), a small lake adjacent to the site. Both water and sediment samples were taken from the lake. The mill tailings can leach directly into both Iso Hiislampi and the stream flowing on its eastern side. Therefore, soil samples were taken between the tailings and the lake (location 8) and between the tailings and the stream (location 9). Samples from the tailings themselves were also taken. Soil samples and run-off sediment samples near the waste-rock pile, water samples (the stream starting from the run-off sediment was dry), lake sediment samples and soil samples between tailings area and the lake were collected in autumn 2010. Samples from the tailings, water samples (the stream starting from the run-off sediment

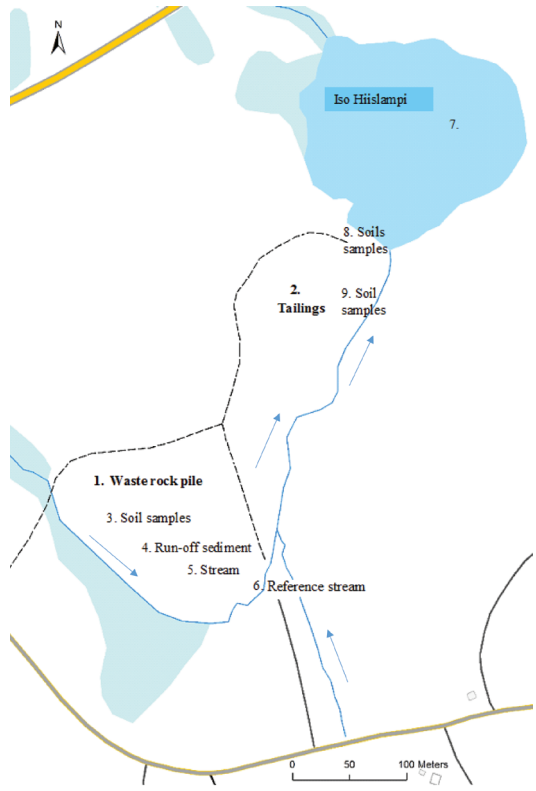


**Fig. 2**. Sampling locations at the former Paukkajanvaara uranium mine. Blue areas indicate wetlands and a lake.

flooded), lake sediment sample and soil samples between the tailings area and the stream were collected in spring 2012.

Three sediment samples were collected from the bottom of Iso Hiislampi (depth approximately 14 m) using a Limnos sediment sampler. The samples were then sliced into 1-cm-thick layers, sealed in plastic boxes and frozen. Soil samples were collected using a topsoil drill and divided into sub-samples on site according to their content. Fifteen samples were collected at the mining site: 5 near the waste-rock pile, 6 between the tailings area and Iso Hiislampi and 4 between the tailings area and the stream. Eight run-off sediment samples were also collected near the waste-rock pile. Three core samples of tailings from variable depths were collected by the Geological Survey of Finland using a drill rig (GM50). Eleven water samples were collected including two reference samples from a small pond just outside the mining area (reference 1) and from a stream that crosses

the Paukkajanvaara site but with no connection to the waste-rock pile (reference 2). Water samples of 10 ml were filtered on site with 0.45-$\mu$m polypropylene filters to pre-treated tubes containing 0.05 ml of concentrated $HNO_3$.

The lake sediment samples were dried in the laboratory using a vacuum drier (Christ Alpha 1–4 LSC). Other solid samples were dried at 40 °C and samples containing organic matter at 60 °C for one week. They were sieved to 2 mm to separate pebble from coarse sand except for the lake sediment samples which were already of sufficiently small grain size. The samples were digested in concentrated $HNO_3$ in a MARS 5 microwave prior to determination of uranium and thorium by inductively coupled mass spectrometry (ICP-MS). $HNO_3$ was chosen based on comparative digestion experiments performed previously on the similar materials (Tuovinen *et al*. 2015). The acids used were all suprapur grade (Merck), suitable for ICP-MS. After cooling, the digest was filtered through a 0.45 $\mu$m polypropylene filter. The microwave digests were stored in Nalgene containers in the refrigerator for further analysis.

### Analytical methods

Uranium and thorium concentrations were determined by Agilent 7500 ce/cx ICP-MS, in the case of the solid samples after microwave digestion with $HNO_3$. The resulting spectra were analysed using Masshunter spectral analysis software. Elemental standard solutions were prepared from multielement standard solution (Merck), diluted with 5% $HNO_3$. Water used in preparation of dilute $HNO_3$ was purified using a Milli-Q water purification system (Millipore, USA). For quality assurance, two reference water materials and two reference sediment materials were included in the ICP-MS analysis: Environment Canada, Certified Reference Materials, trace element fortified calibration standard, TM-27.3, lot 0510 and TMDA-64.2, lot 1010; National Water Research Institute, Certified Reference Materials, HR-1 Humber River sediment for Trace Elements, Lot number 001 and Canadian Certified Reference Materials Project and Lake Sediment Reference LKSD-4 lot number 1563. Deviations of the mean value from the listed reference values are less than 5% for the water standards, less than 10% for lake sediment reference sample LKSD-4, and less than 20% for river sediment reference sample HR-1.

Gamma spectrometry was used for determination of natural uranium and thorium series radionuclides from dried samples. $^{238}U$ was measured using the gamma emission of $^{234m}Pa$ at equilibrium with $^{238}U$. $^{226}Ra$ was determined from the progeny nuclides $^{214}Pb$ and $^{214}Bi$ assumed to be in secular equilibrium with their progenitor. In order to prevent the escape of radon and thus ensure the attainment of radioactive equilibrium between $^{226}Ra$ and $^{214}Pb/^{214}Bi$, the samples were sealed in vacuum bags for three weeks. The samples were then counted on a Canberra BE5030 HPGe detector (FWHM 1.77 eV at 1332.5 keV). Prior to analysis, the detector was calibrated with a mixed nuclide standard (NPL) of similar geometry and density (washed sand, Thermo Fisher). The spectra were analysed using Gamma99 spectral analysis software.

## Results

### Release of U series radionuclides from the waste-rock pile

#### Soil samples

To study leaching of radionuclides and metals from the waste-rock pile, five soil profiles were taken from its southeastern edge (location 3 in Fig. 2) approximately 5 m apart. The first sample SO-1 was taken near the location where the first run-off sediment sample R-1 was collected.

Activity concentrations (Table 1) of $^{238}U$ in the soil samples at $\geq 0.3$ Bq g$^{-1}$ are typical of uraniferous deposits in the region and around an order of magnitude higher than the mean for Finnish soils (0.03–0.06 Bq g$^{-1}$; Sillanpää *et al*. 1989). Values resemble those found at Riutta (0.1–0.8 Bq g$^{-1}$), situated 12 km from Paukkajanvaara, which was used as a reference site by the Finnish Radiation and Nuclear Authority during the mine remediation programme (Lemmelä 1984, Mustonen *et al*. 1989). In contrast to Riutta, there is very substantial disequilibrium

in the uranium series; $^{226}$Ra in all samples was enriched relative to $^{238}$U, in some cases by a substantial amount, and at all soil depths. Concentrations of $^{210}$Pb was lower than that of $^{226}$Ra owing to the incomplete ingrowth of $^{210}$Pb.

### Run-off sediments

The run-off sediment samples were taken at 10 m intervals from a dry stream bed near the waste-rock pile, starting from the beginning of the stream (between locations 4 and 5; *see* Fig. 2).

The values correspond to those determined for soil samples taken from the southeastern edge of the waste-rock pile with $^{238}$U varying from approximately 0.5 Bq g$^{-1}$ (sample R-8) to 3 Bq g$^{-1}$ (sample R-2) (*see* Table 2). The highest activity concentrations occurred 10 m from the start of the stream and generally decreased in the flow direction. In all samples, $^{226}$Ra was enriched relative to parent $^{238}$U, and $^{210}$Pb depleted owing to incomplete ingrowth.

### Water samples

Nine water samples were collected from a stream (Fig. 2) crossing the site to determine possible leaching of uranium from the waste-rock pile. Sample W-1 was taken from the beginning of the stream; thereafter, the samples were taken approximately 40–100 m apart. Sample W-5 was taken from a small pool of standing water approximately halfway along the flow path. The stream flows to Iso Hiislampi, from where sample W-9 was taken. Sampling locations W-1 to W-6 were located upstream of the tailings area.

Overall, U concentrations (Fig. 3) greatly exceed the mean for Finnish streams (0.07–0.21 $\mu$g l$^{-1}$, *see* Tenhola and Tarvainen 2008), as would be expected for a mine site. With the exception of W-5, water samples collected upstream of the tailings area contained much less uranium (2–4 $\mu$g l$^{-1}$) than those affected by the tailings. Sample W-5 was taken from a small pool of stagnant water which seemed to act as a local sink and accounted for the anomalously high value found. Uranium concentrations downstream reached a maximum at W-8 (Fig. 3). The concentration of uranium in Iso Hiislampi surface water sample was similar to that in the stream water in the discharge zone.

## Release of radionuclides from tailings

### Tailings

Three core samples were collected from the tailings at depths of up to 5 m (location 2 in Fig. 2). The samples consisted of sandy deposits with the exception of sample MT-1, where an organic layer of ~3 cm thickness occurred at 4 m; this was the original peat on top of which the tailings were placed.

**Table 1**. Mean ± SD ($n$ = 3) activity concentrations (Bq g$^{-1}$) of $^{238}$U, $^{226}$Ra and $^{210}$Pb in soil samples taken between the waste-rock pile and run-off sediment at Paukkajanvaara.

| Sample | Depth (cm) | U-238 | Ra-226 | Pb-210 |
|---|---|---|---|---|
| SO-1 | 0–3 | 0.5 ± 0.1 | 1.5 ± 0.3 | 0.8 ± 0.2 |
| | 3–10 | 0.6 ± 0.1 | 1.1 ± 0.2 | 0.4 ± 0.1 |
| | 10–20 | 0.3 ± 0.1 | 0.6 ± 0.1 | 0.2 ± 0.1 |
| SO-2 | 0–3 | 1.0 ± 0.2 | 2.3 ± 0.3 | 1.0 ± 0.3 |
| | 3–10 | 1.3 ± 0.2 | 2.3 ± 0.3 | 1.1 ± 0.3 |
| | 10–18 | 0.8 ± 0.1 | 1.7 ± 0.2 | 0.9 ± 0.2 |
| SO-3 | 0–3 | 1.36 ± 0.02 | 3.4 ± 0.5 | 1.9 ± 0.5 |
| | 3–9 | 3.0 ± 0.4 | 6.5 ± 0.7 | 2.7 ± 0.7 |
| SO-4 | 0–3 | < 0.01 | 1.1 ± 0.3 | 0.6 ± 0.2 |
| | 3–13 | 0.26 ± 0.05 | 0.8 ± 0,1 | 0.3 ± 0.1 |
| SO-5 | 0–3 | < 0.01 | 0.4 ± 0.1 | < 0.01 |
| | 3–11 | < 0.01 | 0.3 ± 0.1 | < 0.01 |

**Table 2**. Mean ± SD ($n$ = 3) activity concentrations (Bq g$^{-1}$) of $^{238}$U, $^{226}$Ra and $^{210}$Pb in run-off sediment samples taken from a dry stream bed near the waste-rock pile.

| Sample | U-238 | Ra-226 | Pb-210 |
|---|---|---|---|
| R-1 | 0.6 ± 0.1 | 0.9 ± 0.1 | 0.6 ± 0.2 |
| R-2 | 2.9 ± 0.4 | 6.2 ± 0.7 | 2.7 ± 0.7 |
| R-3 | 1.6 ± 0.2 | 2.9 ± 0.3 | 1.2 ± 0.3 |
| R-4 | 2.2 ± 0.3 | 3.5 ± 0.5 | 0.8 ± 0.2 |
| R-5 | 1.8 ± 0.3 | 2.7 ± 0.4 | 0.7 ± 0.2 |
| R-6 | 0.9 ± 0.1 | 1.81 ± 0.03 | 0.4 ± 0.1 |
| R-7 | 1.9 ± 0.3 | 3.5 ± 0.4 | 0.6 ± 0.2 |
| R-8 | 0.4 ± 0.1 | 0.6 ± 0.1 | 0.3 ± 0.1 |

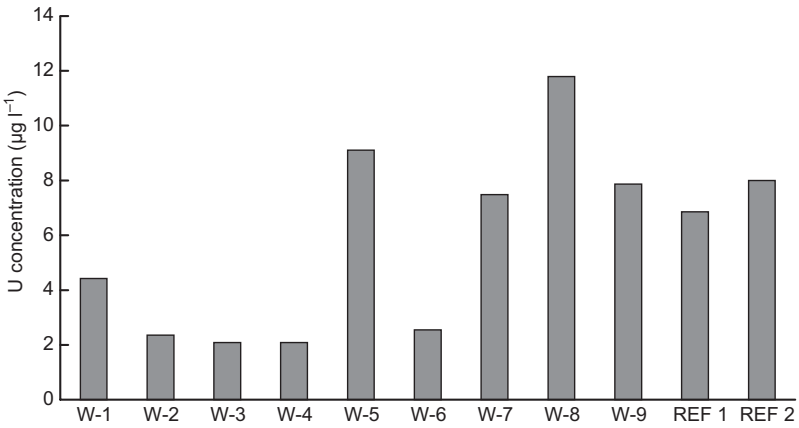


**Fig. 3**. Uranium concentrations in stream-water samples.

The highest concentrations of $^{238}U$ and $^{226}Ra$, 36 400 Bq kg$^{-1}$ and 30 400 Bq kg$^{-1}$, respectively, were found in sample MT-1 from 4 m depth in the organic layer (Table 3). $^{210}Pb$ could not be detected at this depth. Below the organic layer, the concentrations were lower: 448 Bq kg$^{-1}$ for $^{226}Ra$ and 565 Bq kg$^{-1}$ for $^{238}U$. The $^{226}Ra/^{238}U$ ratios were close to unity for tailings samples MT-1 and MT-2 varying from 0.7 to 1.1. In sample MT-3 from 3–4.5 m depth, the absence of $^{238}U$ could be seen clearly; and at 5 m depth, the radionuclide ratio isotopic ratio was 0.6. The radionuclide ratio of $^{226}Ra/^{210}Pb$ varied throughout the sampling depths from 1.1 to 2.0; all the ratios being > 1.

## Soil samples

Six soil samples were collected between Iso Hiislampi and the tailings area (location 8 in Fig. 1). The U and Th concentrations (Fig. 4) in each layer of sample SO-6 were the highest throughout; from 52 mg kg$^{-1}$ in the bottom mineralized layer to 311 mg kg$^{-1}$ in near-surface organics. The highest concentration found in other samples was 78 mg kg$^{-1}$ in the mineralized zone of SO-8. Uranium concentrations in the respective organic layers were highest in samples SO-6, SO-9 and SO-10, indicating surface leaching of uranium from the source and uptake by organic matter. Levels in the other samples SO-7–SO-11 were similar to those found at the Riutta reference site (8–65 mg kg$^{-1}$).

Four soil samples were collected between the stream and tailings area (location 9 in Fig. 2). Activity concentrations of U and progeny in these soil samples were much lower (Table 4) than found in the five soil samples taken between the waste-rock pile and run-off sediment (Table 1). Moreover and in contrast to the latter, there was no enrichment in $^{226}Ra$. In the organic layer of sample SO-12, $^{210}Pb$ was below

**Table 3**. Activity concentrations of $^{238}U$, $^{226}Ra$ and $^{210}Pb$ (Bq kg$^{-1}$) and the ratios of $^{226}Ra/^{238}U$ and $^{226}Ra/^{210}Pb$ in drill cores (diameter 3.5 cm) through tailings deposits at Paukkajanvaara (measurement uncertainty approx. 7%).

| Sample/depth | U-238 | Ra-226 | Pb-210 | $^{226}Ra/^{238}U$ | $^{226}Ra/^{210}Pb$ |
|---|---|---|---|---|---|
| MT-1/2.5 m | 28 | 30 | 19 | 1.1 | 1.6 |
| MT-1/4 m (waste rock) | 2450 | 2590 | 2260 | 1.1 | 1.1 |
| MT-1/4 m (organic layer) | 36400 | 30400 | < 10 | 0.8 | – |
| MT-1/4.5 m | 565 | 448 | 227 | 0.8 | 2.0 |
| MT-2/3 m | 305 | 345 | 227 | 1.1 | 1.5 |
| MT-2/3.5 m | 4980 | 3670 | 2770 | 0.7 | 1.3 |
| MT-3/3 m | < 10 | 26 | < 10 | – | – |
| MT-3/4 m | < 10 | 31 | 26 | – | 1.2 |
| MT-3/4.5 m | < 10 | 27 | < 10 | – | – |
| MT-3/5 m | 2510 | 1490 | 897 | 0.6 | 1.7 |

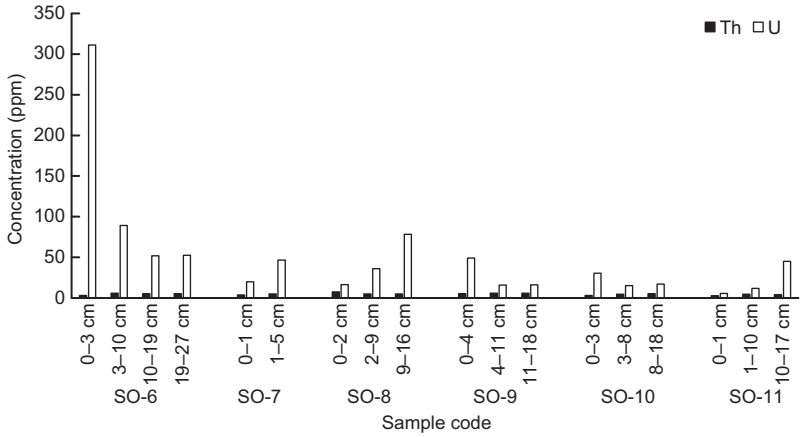


**Fig. 4**. Uranium concentrations in soil samples taken between Iso Hiislampi and the mill tailings area.

the detection limit (approx. 0.01 Bq $g^{-1}$), which indicates its incomplete ingrowth and the leaching of $^{238}U$ and $^{226}Ra$ with the surface flow and binding to organic matter. The same was found in sample SO-13 in both the organic and mineral layer. Samples SO-14 and SO-15 were enriched in $^{210}Pb$ at the surface. This may have been caused by leaching of $^{238}U$ and $^{226}Ra$ and/or settling of $^{210}Pb$ after $^{222}Rn$ decay in the atmosphere. At the subsurface (2–11 cm) in sample SO-14, $^{238}U$ and $^{226}Ra$ were in equilibrium. $^{210}Pb$ could not be detected. This again indicates leaching of $^{238}U$ and $^{226}Ra$ with the surface flow and incomplete ingrowth of $^{210}Pb$.

## Concentration of radionuclides in Iso Hiislampi sediments

Three bottom sediment cores were collected from the centre of Iso Hiislampi and sliced into 1-cm-thick slices immediately after sampling. The upper 6 cm consisted solely of organic matter which is a typical material for lakes such as Iso Hiislampi. At 6–22 cm depth, sediments consisted of either grey sludge, black organic material or a mixture of these and smelt strongly of sulphur. The average sedimentation rate in Finland is 1.5–3.5 mm per year. Assuming an average rate of 2.5 mm per year would imply that the upper 6 cm has formed since the early 1990s; after the remediation operations. The Finnish Nuclear and Safety Authority concluded that sediments from the years 1963–1964 occurred at a depth of 14–16 cm (~30 cm at the date of this study assuming a constant sedimentation rate) and that tailings material started to spread to Iso Hiislampi after the operation ceased (*see* Mustonen *et al*. 1989). This would mean that all the lower layers (> 6 cm) in our study contained a component of tailings and this was clear from both their appearance and the analyses described below.

$^{238}U$, $^{226}Ra$ and $^{210}Pb$ reached their peak concentrations at 7–8 cm, in the layer immediately below the organic-rich deposits (Fig. 5). This together with the very high activity concentrations (80 and 58 Bq $g^{-1}$ for $^{210}Pb$ and $^{226}Ra$, respectively) strongly suggests that waste materials spread directly into the lake before the remediation pro-

**Table 4**. Activity concentrations of $^{238}U$, $^{226}Ra$ and $^{210}Pb$ (Bq $g^{-1}$) in the soil samples collected between the stream and mill tailings (measurement uncertainty approx. 7%).

| Sample | Depth (cm) | U-238 | Ra-226 | Pb-210 |
|---|---|---|---|---|
| SO-12 | 0–1 | 0.09 | 0.09 | < 0.01 |
| | 1–7 | 0.08 | 0.06 | 0.04 |
| | 7–21 | 0.04 | 0.02 | < 0.01 |
| SO-13 | 0–2 | 0.06 | 0.10 | < 0.01 |
| | 2–12 | < 0.01 | 0.02 | < 0.01 |
| SO-14 | 0–2 | < 0.01 | 0.2 | 0.3 |
| | 2–11 | 0.5 | 0.5 | < 0.01 |
| | 11–21 | < 0.01 | 0.02 | 0.02 |
| SO-15 | 0–4 | < 0.01 | 0.3 | 0.6 |
| | 4–8 | 0.09 | 0.10 | 0.07 |
| | 8–22 | < 0.01 | 0.03 | < 0.01 |

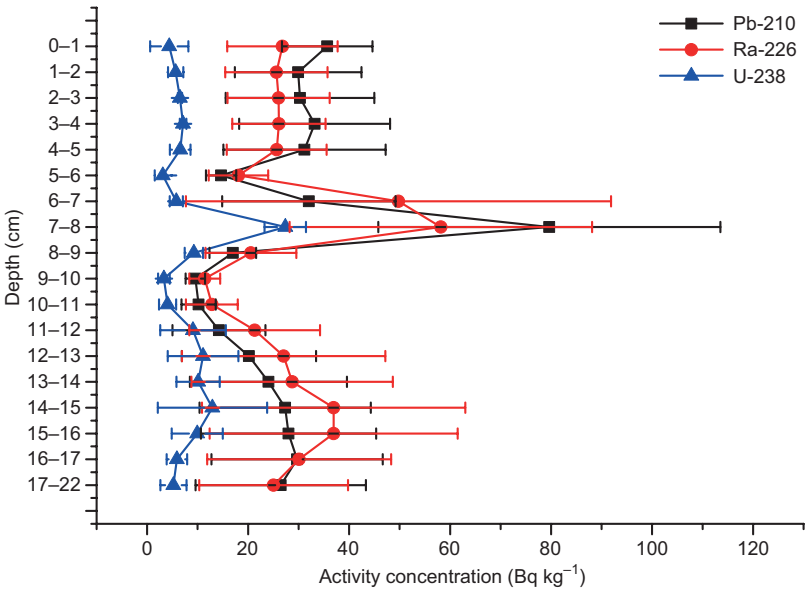


**Fig. 5**. Mean ± SD ($n$ = 3) activity concentrations of $^{238}$U, $^{226}$Ra and $^{210}$Pb as a function of depth in three bottom sediment samples from Iso Hiislampi.

gramme. Further, the enrichment of progeny isotopes over $^{238}$U implies that the material comprised tailings rather than waste rock.

Uranium series disequilibrium provides evidence for subsequent mobilization of the respective isotopes within the sediment column. The variation in the uranium concentrations above (6–7 Bq g$^{-1}$) and below (3–12 Bq g$^{-1}$) with a peak at 7–8 cm could be solely attributed to changes in the physical recharge; chemical mobilization of U as U(VI) would not be expected under the strongly reducing conditions encountered. Radium is also redox sensitive owing to the influence of the sulphur speciation on the alkali earth metal solubility. Interestingly, $^{226}$Ra increased almost monotonically with depth in the lower sediments (8–16 cm), corresponding to progressively more reducing conditions under which precipitation of sulphates would be repressed. In each of those layers, $^{210}$Pb was depleted relative to $^{226}$Ra. By contrast, sediments above the peak (< 6 cm) had fairly constant activity concentrations of $^{226}$Ra (approximately 30 Bq g$^{-1}$) and were enriched in $^{210}$Pb.

## Discussion

Soil and run-off sediment samples were taken to study releases of $^{238}$U and $^{226}$Ra and $^{210}$Pb from the waste-rock pile at Paukkajanvaara. Water samples were also collected from the nearby stream. The concentrations of $^{238}$U and $^{226}$Ra and $^{210}$Pb are considerably higher in the soil samples taken near the southeastern edge of the waste-rock pile than at the nearby Riutta reference site or in Finland on average (Mustonen *et al*. 1989). The radionuclide ratios of the soil samples suggest that $^{226}$Ra has accumulated in surrounding soil. Frissel and Koster (1990) reported that, in normal soil, $^{226}$Ra is almost immobile, but an increase in the soil acidity may enhance its mobility. At Paukkajanvaara, the cause for the increase in the acidity might be caused by oxidation of pyrite grains. The complete oxidation of pyrite is described ains West *et al*. (1991).

In the case of uranium, as the conditions become more oxidizing, uranous species are oxidized to uranyl species $UO_2^{2+}$ (Langmuir 1978), in which uranium is in its more mobile +VI state.

In the run-off sediment samples collected from a dry stream bed near the waste-rock pile, the activity concentrations of $^{238}$U and $^{226}$Ra are slightly higher than in the soil samples. The radionuclide ratios of $^{226}$Ra/$^{238}$U and $^{210}$Pb/$^{226}$Ra correspond to the ratios determined for the soil samples. In all samples, $^{226}$Ra is enriched relative to parent $^{238}$U and $^{210}$Pb depleted owing to incomplete ingrowth. In the water samples collected between the run-off sediment towards Iso

Hiislampi, the average concentration of uranium exceeds the average concentrations in Finnish streams, as would be expected for a mine site. The concentration increased after contact with the tailings deposit towards the lake.

The other contamination source was the tailings, from which radionuclides could leach to both the lake and the stream flowing on its eastern side. Therefore, further soil samples were taken between the tailings area and the lake, and between the tailings area and the stream. In addition, tailings samples were taken. In tailings samples MT-1 and MT-2, the $^{226}Ra/^{238}U$ ratios were close to unity varying from 0.7 to 1.1. Values > 1 would be expected, since uranium was leached from the ore using sulphuric acid during the mining operations, and most of $^{226}Ra$ remained undissolved in the leaching process (Lottermoser 2007). This might indicate that tailings samples partly represented material which had been treated only physically without hydrometallurgical treatment. Any further conclusions cannot be made because there are no documents available regarding original composition of tailings. Only in sample MT-3 from 3–4.5 m depth, deficiency of $^{238}U$ could be seen. At 5 m depth, the $^{226}Ra/^{238}U$ ratio was again 0.6; clearly < 1, which indicates the leaching of $^{238}U$ from upper layers and/or leaching of $^{226}Ra$ to lower layers in the deposit.

The radionuclide concentrations in soil samples collected between the tailings area and Iso Hiislampi showed no clear evidence of the leaching of uranium and thorium to this direction. In the case of the samples collected between the tailings area and the stream, the results indicate leaching of $^{238}U$ and $^{226}Ra$ with the surface flow. The concentrations were, however, much lower than found in the five soil samples taken between the waste-rock pile and run-off sediment.

The stream passing through the Paukkajanvaara mine area flows into Iso Hiislampi. In the sediment cores taken from the bottom of the lake, peak concentrations of $^{238}U$, $^{226}Ra$ and $^{210}Pb$ were found in a layer dated to be coeval with the time of remediation. The peak concentrations in the lake sediment samples most probably represent disposal of fine-grained tailings material to the lake. In the upper organic-rich layers of the core (< 6 cm), the concentrations of $^{226}Ra$ were fairly constant and the layers were enriched in $^{210}Pb$, which might have been caused by the settling of $^{210}Pb$ after $^{222}Rn$ decay in the atmosphere, mobilization of $^{226}Ra$ to lower layers in the sediment profile and/or leaching of $^{210}Pb$. The concentrations of $^{226}Ra$ as compared with those of $^{238}U$ also suggest that there was further leaching of $^{226}Ra$ from the waste-rock pile and/or tailings area.

## Conclusions

Based on the results presented in this paper, at least $^{226}Ra$ was leaching from the waste-rock pile and/or tailings repository to surrounding soil and further to the run-off sediment. Also uranium was enriched in this sediment. During spring floods, these nuclides emanated further in the stream. Increases of uranium concentrations could also be detected after the stream had passed the tailings area. There was also evidence for vertical leaching of $^{226}Ra$ in the tailings repository and the analytical results from the soil samples collected between the repository and the stream suggest that this might be the possible contamination pathway. Finally, the leakage of $^{226}Ra$ could be seen in the top layers of the Iso Hiislampi sediments. Therefore, our study shows that leaching and accumulation of $^{226}Ra$ from the waste-rock pile and possibly tailings at Paukkajanvaara was still an ongoing process.

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